THE Z-SCAN TECHNIQUE TO CHARACTERIZE NON LINEAR OPTICAL MATERIALS: MODELING AND EXPERIMENT

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ABSTRACT

I treat the Z-scan technique applied to characterize a non linear material, probed by a laser beam on one of its atomic transitions. By assuming the caustic of the probing optical beam in accordance with the geometric optics approximation, I model the Z-scan setup. I describe a Z-scan experiment I performed in CDTA on a ruby crystal sample with a top hat laser beam from a Nd:YAG laser followed by a second harmonic generator. The experimental and theoretical Z-scan curves, thus obtained, show good agreement. The cubic non linearity in the material sample, ruby, is eventually determined.

Keywords: Z-scan Technique; theoretical modeling; Z-scan experiment, Non linear optical material.

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1. INTRODUCTION

The invention of laser has allowed the discovery of the non lineare behavior of optical materials made possible by the analysis of the material interaction with high concentrations of optical power fluxes. Nowadays, non linear optical materials find diverse applications. As examples [1]

- The generation of the higher harmonics of laser beam frequencies allowing laser emission at other wavelengths than that possible from the active laser material.
- The shift of laser emission upwards to wider spectra as it is the case for the optical parametric oscillators (OPO) giving yield for many applications, especially in spectroscopy.

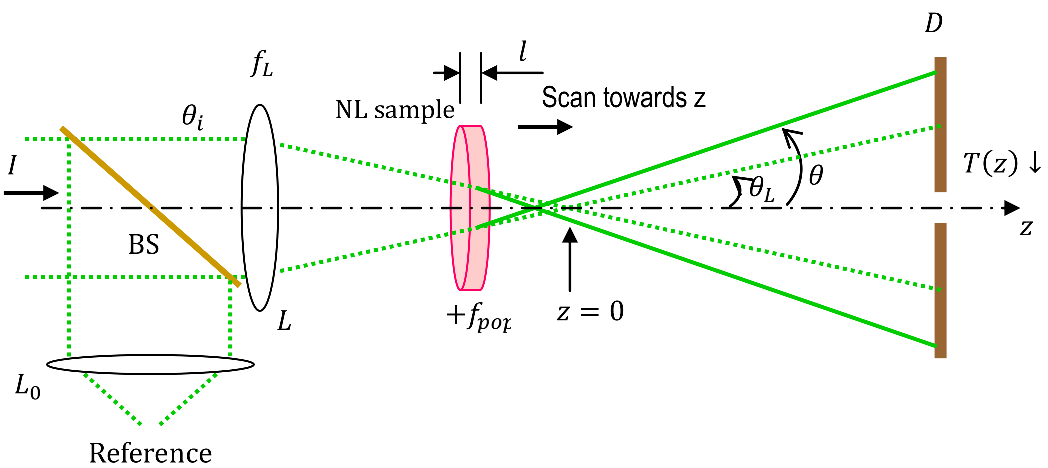
- Dispersion compensation in optical fibers, of paramount importance in optical communications, particularly by generation of propagating solitons.
- The optical switching, the optical computations, etc.

Neverthless, to access these applications, the used materials should have been completely characterized with respect of their non linear behavior. The Z-scan propose by Sheikh-Bahae et al. [2] is the most direct and efficient technique among those used so far [3-5]. It gives acess to both the sign and amplitude of the optical non linearity [6]. Its sensitivity allows the probing of as weak phase shifts as at $\lambda/10^4$ [7].

In this contribution, I describe in section two the Z-scan procedure and derive in section three an analytical formula that depicts its known curve. In section four I describe a Z-scan experiment I performed in CDTA on the laser material ruby by probing it with a top-hat beam from the neodymium YAG laser. Agreement between theoretical and experimental results allows one to deduce the polarizability of ruby as an example of a non linear (NL) optical material.

2. THE Z-SCAN TECHNIQUE

The Z-scan setup is illustrated in Fig. 1. The optical beam from a laser is splitted by means of



a)

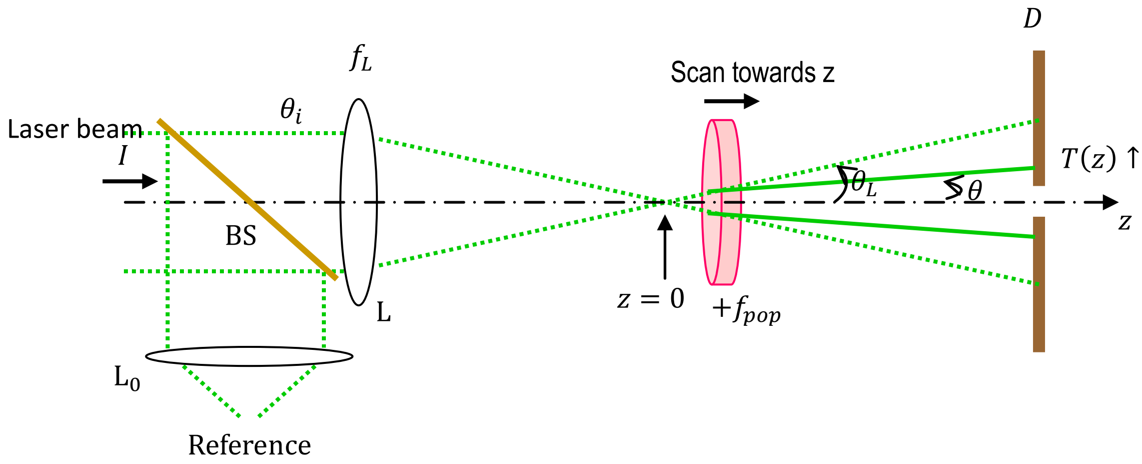


Fig.1. The Z-scan setup, necessating only a beam splitter BS, two lenses L_0 and L and a smaple from a non linear optical material as well as a diaphragm D. The optical mountings that hold the optical elements should use micrometric translators for precise alignment and fine stepping scan. Assuming a positive lensing effect in the NL sample,

a) the beam transmittance $T(z)$ through the diaphragm D decreases as the NL sample scans before the focus of lens L.

b) $T(z)$ increases as the NL sample scans after the focus of lens L.

a beam splitter BS into two components: a reference component measured in the reference branch of the Z-scan setup. The other, horizontal, is the signal branch (Fig.1). The reference component normalizes the signal component passing through the Z-scan lens L and a sample of a NL optical material. The lens L focuses the input beam which is transmitted through a diaphragm D placed in the far field zone of the same focued beam. We quote the transmittance by that diaphragm as $T(z)$ where z is the NL sample position. The latter contains a lensing effect that adds more or less convergence to that beam depending on its non linearity sign. Let's assume a positive lensing effect with a corresponding focal length: $+f$. As the sample scans the focused beam, shown in dotted line (Fig.1-a), before its focus at z = 0, the beam, in continuous line, undergoes more convergence. Then, the transmittance $T(z)$ through D gets less amplitude. Conversely, when the NL sample scans the beam after its focus it causes the beam to get less divergence. Consequently, $T(z)$ through D inreases in amplitude. We see clearly that transmittance T through D is sensitive to the NL sample position. Its behavior depends on the sign of the NL effect in the optical material sample. That is if

- $T(z)$ decreases for $z < 0$
- $T(z)$ increases for $z > 0$

The NL of the optical material is positive: $+f$.

If the contrary occurs, then the NL of the material is negative: $-f$. Between those two situation, at focus where $z = 0$, no focusing effect affects the beam.

The gap between the transmittance maximum (peak) and minimum (valley) relates to the value of the material non linearity. Thus, we see clearly the ability of the Z-scan technique to determine both the sign and value of the material non linearity.

The inventors of the technique, Sheik-Bahae et al., have given empirical formulae [2,3] enabling to find the on-axis phase shift $\Delta\phi_0$ at the beam focus from the given difference ΔT_{p-v} between the normalized peak T_p and valley T_v transmittances by the diaphragm aperture. For a Gaussian transverse profile of the optical beam:

$$\Delta T_{p-v} = 0.406(1-S)^{0.25}|\Delta\phi_0| \qquad (1)$$

S is the interception factor of the beam by the diaphragm aperture. For a Top-Hat profile of the laser beam, one has to apply the formula of Zhao and Palffy-Muhoray [8]:

$$\Delta\phi_0 = 2.7\tanh^{-1}\frac{\Delta T_{p-v}}{2.8(1-S)^{1.14}} \qquad (2)$$

However, the function $T_{p-v}(z)$ is plotted experimentally or computed numerically. $\Delta\phi_0$ in both Eqs. (1) and (2) is the on-axis phase shift caused by the non linearity in the sample material.

3. Z-SCAN MODELING

Let's assume the probing laser beam of Gaussian transverse profile. Then, its on-axis irradiance varies on both sides of the focal region at ranges $z \gg z_R$ (Fig.1) as follows

$$I_0(z) = 2P/\pi w_L^2 = 2P/\pi\theta_L^2 z^2 \qquad (3)$$

P and θ_L are respectively the beam power and divergence after lens L. z_R is the focal depth (Rayleigh range) of that beam, assumed of Gaussien profile. Under optical excitation, the index of refraction of the NL sample writes

$$n = n_0 + \Delta n \qquad (4)$$

n_0 is the linear index and Δn the index change induced optically by the high irradiances. For a cubic non linearity γ (m^2/W), the on-axis index change writes

$$\Delta n_0 = \gamma I_0 \qquad (5)$$

Within the NL sample two lensing effects occur, that of the lens L and the NL one, hence beyond the focal region, their focusing powers add up according to the geometric optics relationship [9,10]

$$\frac{1}{f} = \frac{1}{f_L}\left(1 - \frac{z}{\pm f_{NL}}\right) \qquad (6)$$

$f_L + z$ is the distance separating the two lenses where z is considered in algebric value. The upper sign holds when the lensing effect is positive and vice versa. Eq.(6) shows that the focusing power of the two lensings vary around that of the lens L: $1/f_L$ such that at $z = 0$, the NL lensing loses its effect. This is physically the case as explained above. Taking into account the fact that [10]

$$\frac{1}{f_L} = \frac{\theta_i}{w_{0L}} = \frac{\pi \theta_L \theta_i}{\lambda} \qquad (7)$$

$$\frac{1}{f} = \frac{\theta_i}{w_0} = \frac{\pi \theta \theta_i}{\lambda} \qquad (8)$$

We rewrite Eq.(6)

$$\theta = \theta_L\left(1 - \frac{z}{\pm f_{NL}}\right) \qquad (9)$$

Where λ is the optical beam wavelength, θ_i is the input beam divergence, i.e. before the lens L, θ_L the beam divergence after lens L, assuming that lens acting alone. w_{0L} is the focal width of the beam assuming only the lensing effect of lens L happening. w_0 and θ respectively are the focal width and beam divergence upon combined lensing effects according to Eq. (6).

As in literature [11], let's assume a parabolic radial shape of the refractive index change

$$\Delta n = \Delta n_0 (1 - 2\,\rho^2/w^2) \qquad (10)$$

Where ρ is the radial coordinate and w is the beam width, both in the NL sample. This expression is equivalent to [10]

$$n = n_m (1 - 2\,\rho^2/b^2) \qquad (11)$$

Where

$$b = w\sqrt{\frac{n_m}{\Delta n_0}} \qquad (12)$$

n_m is the maximum on-axis index of refraction. To Eq.(11) corresponds a lensing effect with a focal length [10]

$$f_{NL} = \frac{b}{2n_m \sin(2l_{eff}/b)} \qquad (13)$$

l_{eff} is the effective thickness of the NL sample weighted by the absorbed laser irradiance along its geometrical length l.

$$l_{eff} = \int_0^l e^{-\alpha \acute{z}} d\acute{z} = \frac{1-e^{-\alpha l}}{\alpha} \qquad (14)$$

Here, Fresnel reflection loss at the entrance of the NL sample is ignored so as to consider I_0 the laser irrandiance in the sample volume. α is the absorption coefficient of the NL material.In practice, $n_m \cong n_0$ and $b/w = 10^2$ to 10^3. Counting for thin samples, $l_{eff} \sim w$, then $2\,l_{eff}/b \sim 10^{-2}$ and

$$f_{NL} = \frac{w^2}{4l_{eff}\Delta n_0} \qquad (15)$$

Again, one has to count for the hyperbolic spatial shape of the laser beam, i.e. $w^2 = w_0^2(1 + z^2/z_R^2)$ [12] with

$$z_R = k\,w_{0L}^2/2 = \lambda/\pi\theta_L^2 \qquad (16)$$

(see Eq.7), where $k = 2\pi/\lambda$ is the wave number. Accordingly, using Eqs.(3) and (5), one can rewrite Eq.(9)

$$\theta(\zeta) = \theta_L \left(1 - \frac{8\pi l_{eff}(\pm n_2)tP\zeta/\lambda^2 z_R}{\tau_0(1+\zeta^2)^2}\right) \qquad (17)$$

$\zeta = z/z_R$. As the available laser is pulsed with a Q-switched regime, we considered the transient case by adding a factor t_p/τ_{NL} to the right hand side of Eq.(5). t_p is the pulse temporal length and τ_{NL} is the relaxation time of the nlon linearity.

This formula shows linear dependance of $\theta(\zeta)/\theta_L$ around $\zeta = 0$ and extrema values $\zeta = \pm\sqrt{1/3}$. Their axial separation is $\zeta(\theta_{max}) - \zeta(\theta_{min}) = 1.15$. The transmission through the diaphragm D in Fig.1 writes $T = 1 - e^{-2a_D^2/L^2\theta^2}$, hence

$$T(\zeta) = \frac{1}{S}\left\{1 - \exp\left[\frac{\ln(1-S)}{\left(1-\frac{8\pi l_{eff}(\pm n_2)tP\zeta/\lambda^2 z_R}{\tau_0(1+\zeta^2)^2}\right)^2}\right]\right\} \qquad (17)$$

Where

$$S = T(\theta_L) = 1 - e^{-2a_D^2/L^2\theta_L{}^2} \qquad (18)$$

Is the interception ratio of the beam by the diaphragm aperture of diameter a_D. L being the distance separating the diaphragm from the lens L focus. Again, according to Eqs.(1), (5) et (16), the on-axis phaseshift writes

$$\Delta\phi_0 = \frac{4\pi l_{eff} tP}{\tau_0 \lambda^2 z_R}\gamma \qquad (19)$$

Accordingly,

$$T(\zeta) = \frac{1}{S}\left\{1 - \exp\left[\frac{\ln(1-S)}{\left(1-\frac{2\zeta(\pm\Delta\phi_0)}{(1+\zeta^2)^2}\right)^2}\right]\right\} \qquad (20)$$

In order to determine the phase shift $\Delta\phi_0$ and the NL index of refraction, it is worth approaching the experimental curve by the thoretical curves: (17) or (20).

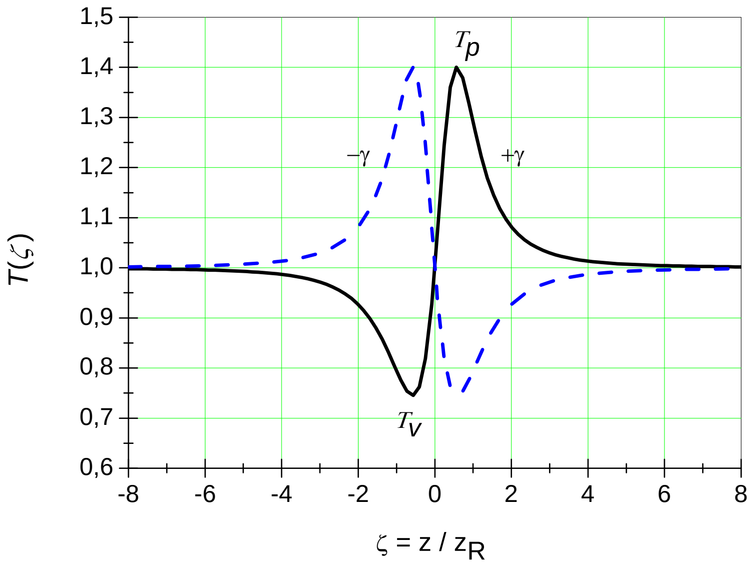
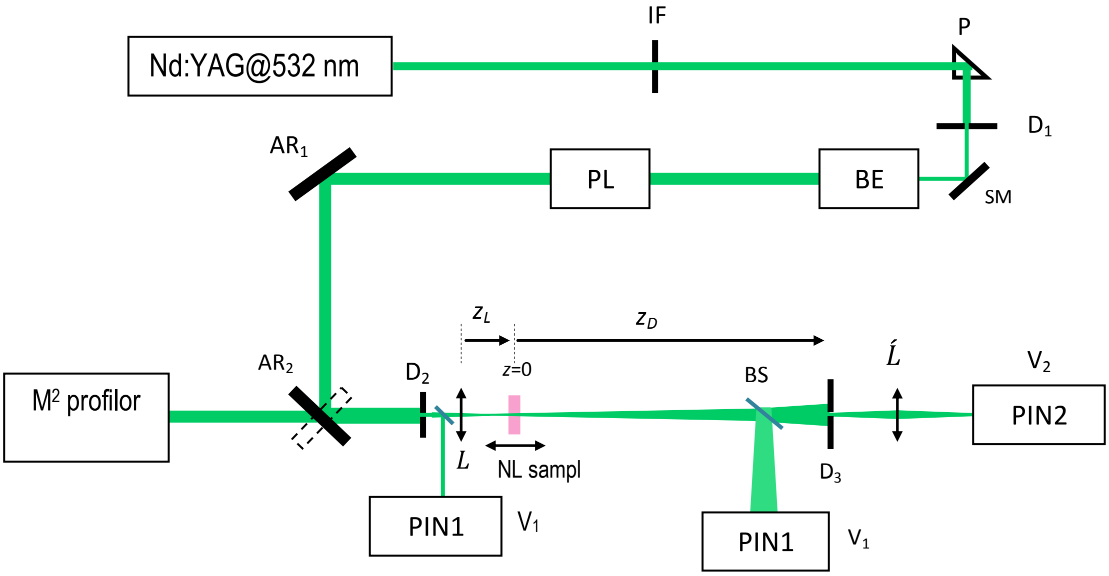


Fig. 2. Plot of the Z-scan transmission (Eq.17) for two non linearities of opposite signs. The numerical parameters considered are: $a_D = 0.5$ mm, $L = 300$ mm, $\theta_L = 5$ mrad, $l_{eff} = 1.5$ mm, $\gamma = 1.25 \times 10^{-8}$ cm^2/W, t_p =10 ns, $P = 7$ kW, $\lambda = 532$ nm, $z_R = 6$ mm, $\tau_0 = 3.6$ ms, S = 20 %.

4. Z-SCAN EXPERIMENT

To experiment the Z-scan, I used a Nd:YAG laser emitting Q-switched pulses @532 nm. The setup is shown in Fig.3. The laser beam is Top-hat with quality factor measured at $M^2 = 1.7$ with a Spiricon beam profiler. In order to take the new beam profile into account, it is enough to replace in Eqs. (17) and (20): ζ, θ_L, z_R, and $\Delta\phi_0$ by $M^2\zeta$, $M^2\theta_L$, z_R/M^2, and $M^2\Delta\phi_0$ respectively. Fig.4 illustrates the experiment curve, in small rounds, obtained on the setup of Fig. 3. The curve in continuous line from Eq. (20) shows good agreement with the experiment measurements. The non linearity sign is positive and its on-axis phase shift is determined at $\Delta\phi_0 = +0.042$. The determination of the non linearity γ is straightforward from Eq.(19) above.



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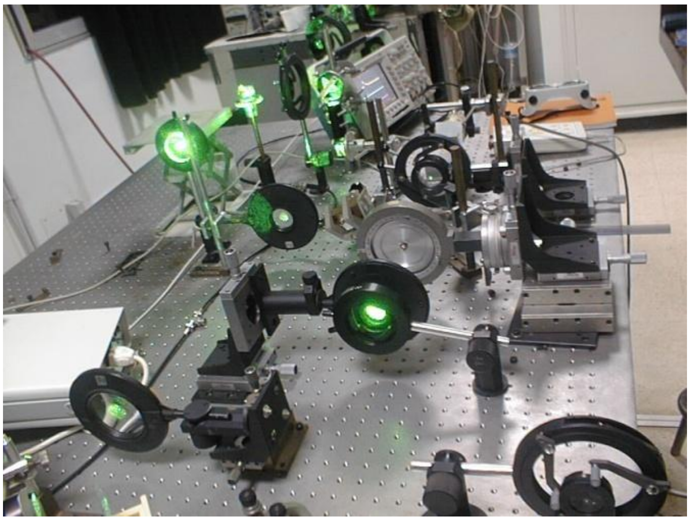

Fig.3. Experimental setup for Z-scan with a Top-hat beam from a Nd :YAG Q-switched laser @53 nm. ***a*)** Experiment schematic with: IF: interfential filtre, D: diaphragm, P: prism, SM: steering mirror, BE: beam expander, (×3), PL: polarizer at Brewster angle, AR_1 et AR_2: aligning mirrors, $\acute{L}$: concentrating lens, L: Z-scan lens with f_L = 100 mm, PIN1 et PIN2: rapid photodiodes in silicon measuring potentials V_1 and V_2, scan range: z_L = 70 mm upto 130 mm, L = 215 mm pour z = 0. BS: beam splitter. ***b*)** photo of the experiment setup in CDTA.

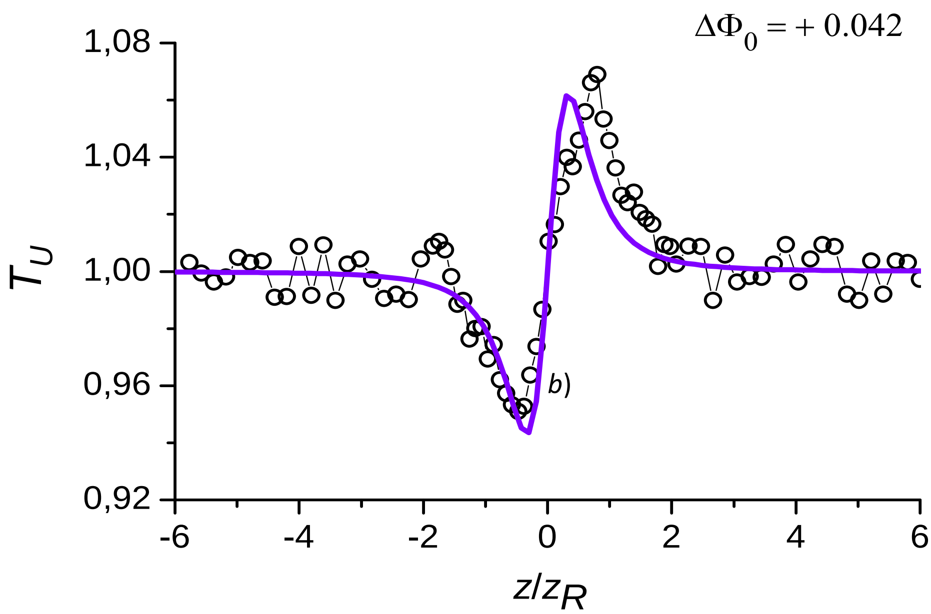

Fig.4. experiment plot (rounds) of the Z-scan transmission $T_U(\zeta)$ where $\zeta = z/z_R$ with a Q-swithced Nd:YAG laser emitting E_0 = 33 ± 3 μJ @ 532 nm with pulse length t_p = 12.4 ns. The lens L with f_L= 100 mm. S = 0.11 ± 0.03, L = 215 mm (Fig.1). The NL sample is made of ruby, a laser material. The continuous line is obtained from Eq.(20).

4. CONCLUSION

In this contribution, I dealt with modeling and experiment of the Z-scan technique for the determination of both the sign and value of a cubic optical nonlinearity in a NL sample. Good agreement is found between the modeling and the experiment demonstrating the merits of both.

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6. REFERENCES

[1] Nie W., Adv. Mater. 1993, 5 (7/8), 520-545, doi: 10.1002/adma.19930050704

[2] Sheik-Bahae M., Said A. A., Van Stryland E. W., Opt. Lett. 1989, 14 (17), 955-957, doi: 10.1364/OL.14.000955

[3] Sheik-Bahae M., Said A. A., Wei T.-H., Hagan D. J., Van Stryland E. W., IEEE J. Quantum Electron. 1990, QE-26 (4), 760-769, doi: 10.1109/3.53394

[4] Azhar S. M., Anis M. Hussaini S. S., Kalainathan S., Shirsat M. D., Rabbani G., Optics and Laser Technology 2017, 87, 11-16, doi: 10.1016/j.optlastec.2016.07.007

[5] Lim G.K., Chen Z.-L., Clark J., Goh R.G.S. Ng W.-H., Tan H.-W., Friend R. H. Ho P. K. H., Chua L. L., Nature Photonics 2011, 5, 554-560, doi: 10.1038/nphoton.2011.177

[6] Traïche M., Godin T., Fromager M., Moncorgé R., Catunda T., Cagniot E., Ait-Ameur K., Opt. Commun. 2011, 284 (7) 1975–1981, doi : 10.1016/j.optcom.2010.12.014

[7] Xia T., Hagan D. J., Sheik-Bahae M., Van Stryland E. W., Opt. Lett. 1994, 19 (5), 317-319, doi : 10.1364/OL.19.000317

[8] Zhao W., Palffy-Muhoray P., Appl. Phys. Lett. 1993, 63 (12), 1613-1615, doi: 10.1063/1.110712

[9] Hecht E. Optique, cours et problèmes. 1980, McGraw-Hill, Chap. 1

[10] Kogelnik H., BSTJ 1965, 44 (3), 455-494, doi: 10.1002/j.1538-7305.1965.tb01672.x

[11] Kogelnik H., Appl. Opt. 1965, 4 (12), 1562-1569, doi: 10.1364/AO.4.001562

[12] Kogelnik H., Li T., Proc IEEE 1966, 54 (10), 1312-1329, doi: 10.1109/PROC.1966.5119